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Impacts of fuel quality on indoor environment onboard a ship: From policy to practice

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ABSTRACT

Environmental considerations, concerning the negative impacts of ship exhaust gases and particles on ambient air quality, are behind the requirements of cleaner marine fuels currently applied in designated emission control areas (ECAs). We investigated the impact of a ship operating on two types of fuel on the indoor air quality onboard. Gaseous and particulate air pollutants were measured in the engine room and the accommodation sections on-board an icebreaker operating first on Heavy Fuel Oil (HFO, 1%-S), and later Marine Diesel Oil (MDO, 0.1%-S). Statistically significant decrease of SO_2, NOx, $PM_{2.5}$ and particle number concentration were observed when the ship was operating on MDO. Due to the higher content of alkylated PAHs in MDO compared to HFO, the concentration of PAHs increased during operation on MDO. The particulate PAHs classified as carcinogens, were similar to or lower in the MDO campaign. Chemical analysis of $PM_{2.5}$ revealed that the particles consisted mainly of organic carbon and sulfate, although the fraction of metals was quite large in particles from the engine room. Principal Component Analysis of all measured parameters showed a clear difference between HFO and MDO fuel on the indoor environmental quality on-board the ship. This empirical study poses a first example on how environmental policy-making impacts not only the primary target at a global level, but also brings unexpected localized benefits at workplace level. The study emphasizes the need of further investigations on the impact of new marine fuels and technologies on the indoor air environments on board.

1. Introduction

The objective of this work was to study the impact of two different ship fuel qualities, Heavy Fuel Oil (HFO) and Marine Diesel Oil (MDO) on the indoor environmental quality (IEQ) on-board the same ship. The urge to decrease emissions of air pollutants to the atmosphere at global and regional levels has led to more stringent requirements for marine bunker fuels and/or exhaust gas cleaning

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technologies. With this respect, several publications present the effects of the new marine fuels on the ambient air quality. This manuscript presents the procedures and results of a unique study where we had the opportunity to perform before-and-after measurements of the indoor environmental quality on a vessel operating first on HFO (1% sulphur content), and later on MDO (0.1% sulphur content). It is the first study of its kind presenting the results of large number of IEQ parameters such as thermal comfort, concentrations of both gaseous and particulate air pollutants, chemical characterization of particles and statistical analysis of the results, as well as a comparison of the results with Occupational Exposure Limits (OEL) and relevant guideline values.

Environmental considerations and increased societal demands have led to the introduction of more stringent regulation of sulfur content in marine fuels (IMO, 2017). The primary goal is to reduce harmful impacts of shipping on the environment by reducing emissions of SO_2 from ships to air, the main target being global and regional reduction of negative health impacts from both primary and secondary pollutants associated with these emissions. Since 2015, vessels operating in designated Sulphur Emission Control Areas (SECA) must run on fuel with a sulfur content of maximum 0.1% (on mass basis). From 2020, the general sulphur content limit for ships operating also outside the SECAs will be reduced from 3.50% (in effect since 2012) to 0.5% (mass by mass). To meet this requirement, ships can either use low-sulfur compliant fuel for main and auxiliary engines and boilers or use approved exhaust gas cleaning systems.

Emissions from ships have been studied with respect to the regulated gaseous pollutants NO_x and SO_x and particles of various sizes, as well as non-regulated pollutants CO, VOC and PAHs, for various types of ship, propulsion, engine configuration and fuel quality. Investigations of particulate matter largely focused on characterization of primary particle emission in ship exhaust (Fridell et al., 2008) and its dependence on type of fuel (Winnes and Fridell, 2009) or engine loads (Anderson et al., 2015), exhaust cleaning equipment (Hallquist et al., 2012), or the effect of maneuvering ships in ports (Winnes and Fridell, 2010). Chemical characterization of particles with respect to elemental composition, organic and elemental carbon as well as well as emissions of NO_x, SO_2 and CO was performed on ships operating the main engine(s) either on Heavy Fuel Oil (HFO) or marine gas oil (MGO) (Moldanová et al., 2009; Moldanová et al., 2013). The effects of fuel change on the emissions of gaseous air pollutants and particles were evaluated on the same ship that changed from HFO with 1% sulfur content, to a residual marine fuel oil (RMB30) with 0.1% sulfur. The SO_2 and PM emissions were reduced by 80% and 67%, respectively (Zetterdahl et al., 2016). Particle mass and number, NO_x and SO_2 from marine engines using three different types of fuels with different content of sulfur were measured on two ships. The type of fuel was shown to affect particle mass but not particle number concentrations. The fuel type also affects the concentration of SO_2 but not NO_x in the exhaust gases (Winnes et al., 2016).

Contrary to the number of scientific articles about ambient air quality and effects on environment and health due to emissions *from* ships, there is surprisingly little known about the air quality *on-board* ships. Two previous studies evaluated thermal comfort in engine room and engine control room (Orosa and Oliveira, 2010; Liu et al., 2011). Another, more detailed study on IEQ on-board two Korean ships was performed with respect to thermal comfort and concentrations of volatile organic compounds (VOC), formaldehyde, CO, CO_2 and PM_{10} (Kim and Lee, 2010). Indoor environmental quality was investigated in a Swedish submarine. The measured parameters included temperature, relative humidity, CO_2, ozone, NO_2, formaldehyde, VOC and particulate matter (Persson et al., 2006). One attempt to study IEQ on-board a ship, in relation to the type of fuel with different sulphur contents, was performed on a cruise ship operating first on HFO (1%-S) and later on ultra-low sulfur hybrid oil (RMB30; 0.1%-S). The study was limited to gaseous air pollutants NO_x, SO_2 and TVOC (Langer et al., 2016).

The indoor environment on-board a ship is a combined occupational and residential environment. The indoor environmental quality (IEQ) is very important for the crew who can spend prolonged periods of time, from weeks to months, on-board, making it a working as well as a living environment. Seafaring is a risky occupation in many other aspects of working life and ensuring good indoor air quality should be an obvious requirement, important for the crew members' health, work efficiency and well-being. Exposure to hazardous substances from the fuel and exhaust is mentioned as one of the occupational risks of seafaring (Oldenburg et al., 2010). Other studies have shown that exposure to exhaust gas from diesel engines increases the risk for cancer, especially for the crews in the engine department (Saarni et al., 2002; Forsell et al., 2007; Kaerlev et al., 2005). Since 2012, diesel exhaust is classified as carcinogenic to humans by the International Agency for Research on Cancer (IARC, 2012a).

2. Materials and methods

2.1. The ship

The study was performed on board the Swedish icebreaker Oden. An icebreaker was chosen since this type of ship is not involved in any cargo operations that could affect the air quality. Icebreaker Oden is a unique ship. It is intended to break ice in the Baltic Sea during the Swedish winters and further serves as a platform for environmental research in the polar regions during summers. The ship was built 1988 in Gothenburg, Sweden. Oden is 108 m long, 31 m wide, with displacement of 13 000 tonnes. It has a diesel-mechanic propulsion system, powered by four Sulzer ZA40S main engines with total power of 24 500 hp, and is equipped with four auxiliary Sulzer AT 25H diesel generators, 1200 kW each. During the first measuring campaign, the main engines were operating on heavy fuel oil (HFO 1%-S) and the auxiliary engines with marine gas oil (MGO). During the second campaign, both the main and auxiliary engines were operating on Marine Diesel Oil (MDO 0.1%-S). The crew normally consists of 20–25 persons.

2.2. The campaigns

The measurements were performed in two campaigns during the summer season. The first was on a smooth and calm voyage over

an open sea from the coastal city of Landskrona in south-western Sweden to Longyearbyen at Svalbard during the time period of 6–18 August 2013. The second campaign was during a voyage from Longyearbyen going north of Svalbard and back to Longyearbyen again during 15–22 July 2016, mostly performing operations in ice. Tracks illustrating the sea voyages are presented in Figs. S1 and S2. Campaigns were performed during summer time when the icebreaker was involved in research operations in the Arctic and not travelling in convoys with other ships close by, whose exhausts could possibly disturb the investigation. We assume that both campaigns had very little impact from air pollution sources other than the ship alone. Fig. S3 shows the prevailing wind direction and wind speed during the voyages.

2.3. The measurements

The sampling sites on the ship were selected to represent both working and living environments for all professional categories on board. In the engine compartment which it is assumed to be affected by engine operations related pollutants and processes and where the highest level of pollutants is expected, sampling was performed in the boiler room and the purifier room. Further, sampling was performed in engine control room, engine office, on the bridge, where the ship is commanded, in the galley, messroom, and in two cabins in the middle of the accommodation section, as representatives for the personal spaces that are expected to be affected by pollutants entering through the ship's ventilation system. The engine compartment and the living compartment are separately ventilated; the fresh air intake is placed on the main deck and on the third deck for the engine compartment and for the living spaces, respectively. Outdoor samples were collected in the bow of the ship at fourth deck approximately 20 m above sea level.

The measured parameters were temperature, relative humidity (RH), concentration of CO_2, air exchange rate (AER), concentration of SO_2, NO_x (NO and NO_2), Total Volatile Organic Compounds (TVOC) including benzene, polycyclic aromatic hydrocarbons (PAHs), and particles as $PM_{2.5}$ and number concentration and size distribution of nanoparticles.

Temperature, RH and concentration of CO_2 were monitored for 24 h at each sampling site using a Wöhler CDL 210 CO_2-logger (Wöhler Technik GmbH, Germany) with a time resolution of 1 min. The resulting values were arithmetic means of these parameters over the sampling period. AER was measured in all the spaces in which the environmental measurements occurred using inert gas (CO_2) injection and following its concentration–time decay (Vaisala GM70 with CO_2-probe GMP222; S/N J2620119), at the beginning of the first (HFO) campaign. Concentrations of SO_2 and NO_x were sampled using IVL diffusive samplers (Ferm and Svanberg, 1998) and analyzed by wet chemical techniques using spectrophotometric methods. The analytical procedures are accredited by the Swedish accreditation agency SWEDAC.

The volatile organic compounds (VOC) were passively collected on Tenax adsorbent medium (Perkin-Elmer tubes). The sampling and analysis followed the ISO 16017-2 standard (ISO, 2003). Details of the analytical procedure for VOC are described elsewhere (Langer et al., 2015). Shortly, the VOCs were thermally desorbed from adsorbent tubes into a gas chromatograph and detected by a mass spectrometer. TVOC were quantified in toluene equivalents. Benzene was quantified using a compound specific response factor.

Formaldehyde was sampled using a passive DSD-DNPH Aldehyde Diffusive sampling Device (Supelco, Bellefonte, PA). The sampling and analytical technique (solvent extraction and high-performance liquid chromatography) generally followed the procedures described in the ISO 16000-4 standard.

PAHs were collected by polyurethane foam (PUF) passive samplers and 32 PAHs were analyzed by GC/MS. Published uptake factors for individual PAHs in gas and particulate phase, respectively, were used as basis for the calculations of concentration levels in this study (Bohlin et al., 2010a). PAHs were collected at three sites during the first (HFO) voyage and at ten sites during the second (MDO) voyage. The passive samplers were preferred to active sampling for PAHs in order to obtain time-integrated samples over the same period of time as the other air pollutants (SO_2, NOx, VOCs and formaldehyde), and in difficult work situations which do not allowed the use of battery-powered sampling pumps (explosion risk).

All the passive samplers in one location were installed and uninstalled at the same time. Filed blanks were employed and the results from respective sampler were corrected for the blanks. Due to complexity of the sampling program, duplicate samples were not deployed.

The samples were flown after the campaigns and analyzed within one week after arrival to the laboratory. The results from all the diffusive samplers reflect the time-integrated mean concentration over sampling time of one week for each sampling site.

Particles $PM_{2.5}$ were collected on pre-weighed Teflon filters (Whatman, TF-1000, 25 mm, pore size 1 μm) and quartz filters (Pall, Tissuequarz, 25 mm); backup quartz filters were used to evaluate and correct for the positive sampling artefact. Impactors made of Teflon using sampling flow rate of 17 L/minute were used for the $PM_{2.5}$ collection (Ferm et al., 2001). Sampling time was ~ 24 h resulting in sampled volume of approximately 25 m^3 of air. The PM mass was determined only on the Teflon filter samples by weighing at constant temperature (23 °C) and RH (50%) conditions, using a microbalance with sensitivity of 1 μg (Sartorius M5P, Data Weighing Systems, USA). The $PM_{2.5}$ was calculated from the particle mass on the filters and the sampled volume. Limit of Detection for the particle weight on a filter was 10 μg.

Particles collected on the Teflon filter were subjected to elemental analysis (elements from sodium (Na) to lead (Pb)) using Energy dispersive X-Ray Fluorescence (Cooper Environmental Services, Beaverton, OR). The quartz filter samples were used for determination of organic carbon (OC) and elemental carbon (EC) by a thermal-optical method (EC/OC analyzer Model 4, Sunset Laboratory, USA) (Bauer et al., 2009) using the EUSAAR_2 temperature protocol as described in Mašková et al. (2015).

Nanoparticle number concentration and size distribution were measured during both campaigns using on-line instruments. During the HFO-campaign, particles in a size range of 5.6–560 nm divided into 32 electric mobility diameters were monitored with an EEPS (Engine Exhaust Particle Sizer, Model 3090, TSI Inc.). During the MDO-campaign a Grimm mini-WRAS (Grimm Model 1371) was used for the particle measurements in the range 10 – 35 000 nm. A time resolution of 1 min was used for both instruments. The

total number concentration of nanoparticles (d < 1 000 nm) was calculated as the averages over a given sampling period. For the calculation of the total number concentrations, the whole range of the EEPS-instruments and the first 19 size channels for the mini-WRAS were used. Both instruments operate on the same principle, electric mobility spectrometry, in the nanoparticle size range and both use corona charger for particle charging. The performance of these two different particle instruments has been evaluated in a separate project involving two Mini-WRAS and one EEPS apparatus. The comparison showed that total number concentrations in the common size ranges measured with the EEPS instrument were by ~40% lower while the two Mini-WRAS instruments differed by 20–30%. Comparison of the particle size distributions measured by the Mini-WRAS and the EEPS is presented in the Supplementary Material (Fig. S4).

Statistical bi-variate analysis was performed using Wilcoxon matched pairs signed rank test based on median values of measured concentrations to investigate differences between the levels of air pollutants on-board when operating on HFO and MDO, respectively.

Principal Component Analysis (PCA) was used to describe and visualize the complex similarities and differences between the samples. PCA is a mathematical projection method, the purpose of which is to reduce the dimensionality of a data set in which there are a large number of interrelated variables, while retaining as much as possible of the variation present in the data. The reduction was achieved by transforming to a new set of variables, the principal components, which are uncorrelated, and ordered so that the first few retain most of the variation present in all of the original variables (Jolliffe, 1986). We have taken the two first components of the PCA result, which thus captures the dominating correlation structure in the data. With two dimensions the data can be visualized in simple scatter plots, one where the observations are pictured, the score plot, and one where the original variables are pictured, the loading plot. The two plots are interrelated as directions in the one plot correspond to directions in the other.

3. Results and discussion

This section reports and discusses the results of the measurements and statistical analyses for the indoor climate parameters and concentrations of the air pollutants from the HFO and MDO campaigns. Occasionally, due to difficulties to set up samples at a measurement site or an analytical misfortune, some data are missing. It is however reasonable to assume that these minor gaps do not affect the overall conclusions of the study.

The typical direction of the wind relative to the ship's movements and thus the fresh air intakes and engine exhaust were similar in both campaigns (Fig. S3). The prevailing northern winds indicated that the wind was blowing from the bow to the aft of the ship (approximately 50% of time) thus leaving the exhaust gases mainly behind the ship. The indoor air quality on-board a ship in the personal spaces may be assessed from the ASHRAE ventilation standard (ASHRAE, 2013) the existing recommended guideline values for good indoor air quality provided by the World Health Organization (WHO, 2005; WHO, 2010), national agencies (UBA, 2019) or recommended in the scientific literature (Mandin et al., 2009). Benzene and benzo(a)pyrene are classified by IARC as carcinogens Group 1 (IARC, 2012b) and naphthalene as a carcinogen Group 2B (IARC, 2002). Since the measurements were carried out on a Swedish flagged ship, the results are compared to Swedish occupational exposure limits (OEL), as regulated by the Swedish Work Environment Authority (AFS, 2008). (AFS, 2018:01)Within the EU, the OEL are the same for SO_2, NO, NO_2 (and naphthalene but the OEL for benzene is 0.5 ppm (1.5 mg/m^3) in Sweden and 1 ppm (3 mg/m^3) in EU, all measured, or in relation to, a reference period of eight hours according to EU Directive 2017/2398, 2017 (EU, 2017). The indoor air guideline values and the occupational limit values for the parameters and compounds measured in this work are summarized in Table S1. The medians of temperature, RH, concentrations of all the air pollutants, as well as p-values from the bi-variate statistical analyses from all the measurement sites are summarized in Table S2.

3.1. Indoor climate parameters.

Indoor climate parameters temperature and relative humidity are important for perception of thermal comfort. The values of temperature and RH at all the measurement sites are shown in Table S3. The differences between median temperature and RH were statistically significant at $p < 0.05$ (Table S2).

There were large differences in temperature and relative humidity between the engine room and the living spaces (Fig. 1). The highest temperature and lowest RH were found in the engine room (average of the boiler and the purifier rooms). In the personal spaces (average of all sites), temperature and RH fell within or slightly outside the comfort zone for good indoor climate: temperature 20–24 °C and RH 30–60%. The outdoor temperature and relative humidity were 13 °C and 76% at the first (HFO) campaign and 2.1 °C and 100% at the second (MDO) campaign.

HFO as fuel requires preheating in all stages of operations – from bunkering to storage in heated tanks, transferring to service tanks, purifying and injection to the cylinders. The boiler and purifier rooms had the largest differences in the temperature and RH since MDO does not require the same preheating for operation or purifying. The temperatures in some places in the engine room were high enough that the crew could experience heat stress during prolonged periods of work. It has been shown that maximum time spent in temperatures around 35 °C should not exceed 35–40 min and after this period of time, relaxation in cooler environments is recommended (Orosa and Oliveira, 2010). The differences in temperature and RH at the other sites on-board are most likely connected to other factors like the temperature and absolute humidity in the outside air rather than the change of the fuel.

Measurement of air exchange rates (AER) at the sites showed that all spaces on the ship were well ventilated. AER in the living spaces were 1.5–10 h^{-1} and between 40 and 80 h^{-1} in the engine room. Recommended AER for dwellings (ASHRAE, 2013) is 0.5 h^{-1}. In this view, the living spaces at the ship were over-ventilated, compared to a home environment. Ventilation rates in the

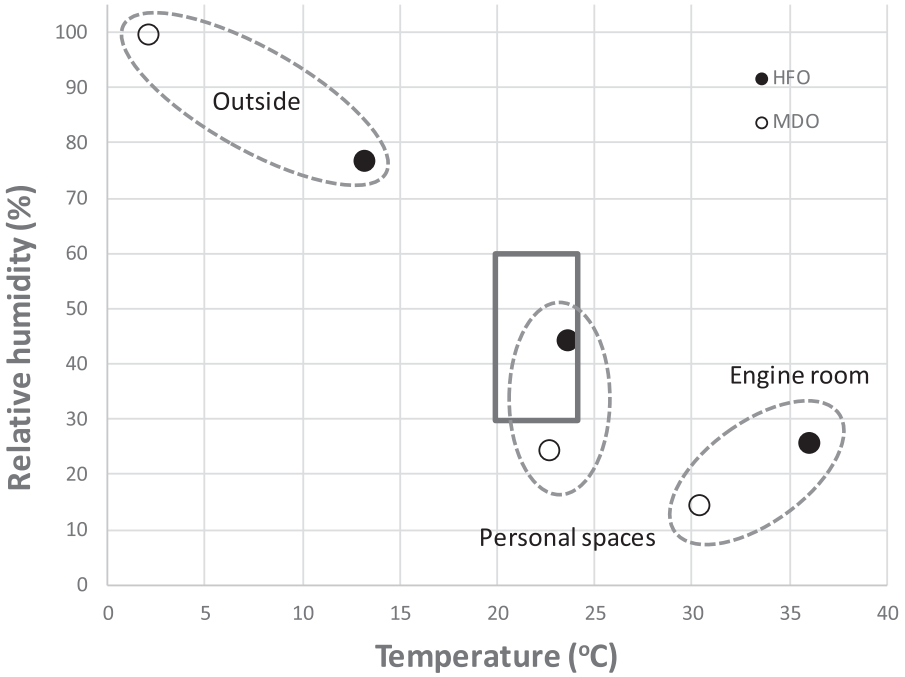

Fig. 1. Plot of relative humidity vs. temperature measured in the ship compartments and outside. The box indicates comfort zone for good indoor environments. Filled circles: HFO, open circles: MDO.

engine room are set for good operation of the engines, ensuring sufficient air flow for efficient combustion. The concentration of CO_2 in all places was close to the ambient CO_2 level, around 400 ppm, thus never exceeding the recommended indoor air guideline value of 1 000 ppm (Table S3).

3.2. Concentrations of SO_2 and NO_x

The median concentrations of SO_2 and NOx as well as those in the individual measurement sites of these air pollutants (Table S4) were all below the indoor air guideline values during both on-board measurements and far below the relevant OEL values (see Table

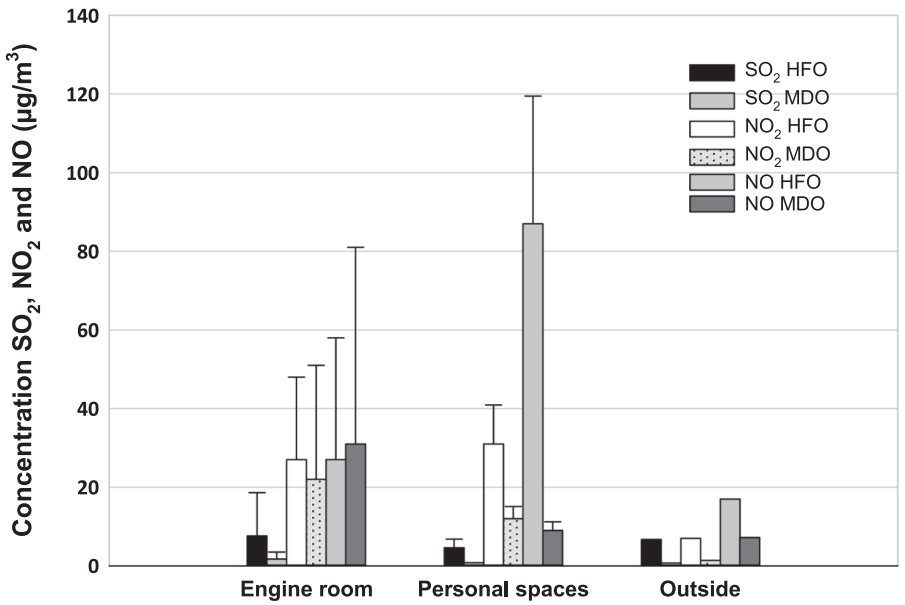

Fig. 2. Concentration of SO_2 and NO_x measured in the ship compartments and outside. The error bars are 95%-confidence intervals except for the outside measurement (single point).

S2).

The largest change between the two campaigns was observed for the concentrations of sulfur dioxide and nitrogen dioxide (Fig. 2). The differences were statistically significant with $p < 0.01$. SO_2 was not detected at levels above the limit of quantification (LOQ) of the method in the case with MDO while concentrations above LOQ were found during HFO operation. Median concentration of nitrogen oxide decreased by almost a factor of 10 but the change was statistically less strong, $p < 0.05$ and the differences may indicate impact from exhaust re-entering the ship interior through the ventilation. The decrease of the concentration of SO_2 is a logical effect of the fuel change to a marine fuel with low sulfur content forced by the SECA regulation (IMO, 2017). These findings are in line with previous studies (Zetterdahl et al., 2016; Langer et al., 2016) where the fuel change on a passenger ferry from HFO (1% S) to ultra-low sulfur hybrid oil (RMB30, 0,1% S) was found to significantly decrease emissions of SO_2 as well as particles.

The concentrations of NO and NO_2 (Table S4) reveal that air quality may be affected by exhaust emissions. The exhaust emissions might enter the ventilation system of the personal spaces from occasional pollution from the exhaust funnel or through mixing in ventilation exhaust from the engine room (less possible). As concentrations of NO_x are high in some compartments and the NO_2/NO ratios are much higher than that of the fresh exhaust (~0.05) but far lower than that of the clean marine troposphere, short processing in the ventilation system or in atmosphere can be assumed.

The sampling site placed outside on the deck also shows higher NO_2/NO ratios indicating influence from a slightly processed (oxidized) exhaust. Comparison of especially NO and NO_2 concentrations indicates that the HFO cruise was more affected by exhaust re-entering the ventilation system than the MDO cruise, as the NO concentrations were similar in the engine room during both cruises while they were higher in the personal spaces, affected by ventilation air, during the HFO cruise.

SO_2 was about five times higher in the engine room during the HFO cruise comparing to MDO. Hence, the measurements in the personal spaces give rather overview of air quality under a range of different conditions while the engine room measurements provide more direct comparison of impact of the two different fuels on indoor air quality.

3.3. Concentration of TVOC and PAHs

The measured values at all the sites are shown in Tables S5–S7. The medians of TVOC, benzene, the sum of 32 PAHs, naphthalene and benzo(a)pyrene as well as p-values from the statistical analyses from all the measurement sites are summarized in Table S2.

Similar to what has been reported in other studies, concentrations of TVOC and benzene vary with type of vessel operations, but also location on-board with the highest concentrations found in the engine room (Kirkeleit et al., 2006; Jacobs et al., 2010; Jacobs et al., 2011). These studies were conducted on tankers and an oil production vessel, which to some extent explains the higher concentrations than the ones observed on the icebreaker. A small decrease in median values of the total concentration of TVOC and benzene was observed when the ship operated on MDO, but these variations are not statistically significant (Table S2).

Tables S6 and S7 show the air concentrations of 32 individual PAHs for all measurement sites for the two sampling campaigns using the PUF passive samplers. All the analyzed PAHs were detected in all samples, indicating that these pollutants are present at the various places on-board. The medians of the sum of 32 PAHs and naphthalene were statistically different ($p < 0.05$) between the two fuels but not for benzo(a)pyrene. The median concentrations of naphthalene and benzo(a)pyrene were below the recommended indoor guideline values as well as the Swedish OEL limit values (Table S1).

Contrary to the other air pollutants, the concentration of PAHs measured as the sum of 32 PAHs increased when the ship was operating on the MDO. The concentrations of naphthalene and benzo(a)pyrene decreased after changing to MDO. The highest level of the sum of 32 PAHs was found in the boiler room (39 000 ng/m^3; MDO fuel) followed by the purifier room (3 300 and 15 000 ng/m^3; HFO and MDO fuel). Somewhat elevated levels were found in the engine control room (2 000 ng/m^3; MDO fuel) and outdoor on the fourth deck (4 300 ng/m^3; MDO fuel). All other places show a lower levels of PAHs on-board (ca 300–1 000 ng/m^3).

The levels in this study were equal to, or higher than typical urban indoor air and background outdoor air situations (Strandberg et al., 2006; Bohlin et al., 2008). Notably, the concentrations found at some of the work places on the ship (boiler, purifier and engine control room) are similar to, or higher than those reported at workplaces where elevated PAH levels are suspected, such as an alloy factory (320–1 900 ng/m^3) (Bohlin et al., 2010b) or among Swedish kitchen workers (85–970 ng/m^3) (Lewné et al., 2017). The elevated PAH levels at the working places on the ship point to the importance to perform careful personal exposure assessment for different ship personal categories.

The PAH patterns in both campaigns were influenced by naphthalene (20–60%), and 3-ringed PAHs (acenaphthalene, acenaphthene, fluorene and phenanthrene; 40–80%). The 5–6-ringed PAHs were low (<2%) although somewhat more prevalent at campaign 1 (HFO fuel). Concentrations of carcinogenic PAH-species included in group of particle-bound PAH species (benzo(a) anthracene to benzo(g,h,i)perylene) were found lower when the ship was operating on MDO.

A comparison of the corresponding locations investigated during the two campaigns (purifier room, cabin and outdoor on the fourth deck) shows that the sum of 32 PAH levels were two to three times higher for the MDO-campaign. This indicates that the PAHs in the fuel affect the environment on the different locations on the ship and, thus, the MDO fuel, compared to HFO causes the highest levels of PAHs on-board.

The environmental impact of the two fuels was also assessed by comparing the PAH pattern in the air samples against measured levels of PAHs in the two fuels (Table S8). There were similarities between the PAH pattern in the air samples from all places at each campaign and the pattern of the PAHs in respective fuel. The reported PAHs in HFO and MDO fuels, respectively, contain ca 75% and 80% of alkylated species. The levels of alkylated PAHs at the measurement sites have a relatively high proportion of these PAHs (40–70%). The significant proportion of the alkylated PAHs in the air emphasize the need to also include these PAHs when performing exposure studies. The higher alkylated proportion at the measurement sites during the MDO campaign compared to the HFO

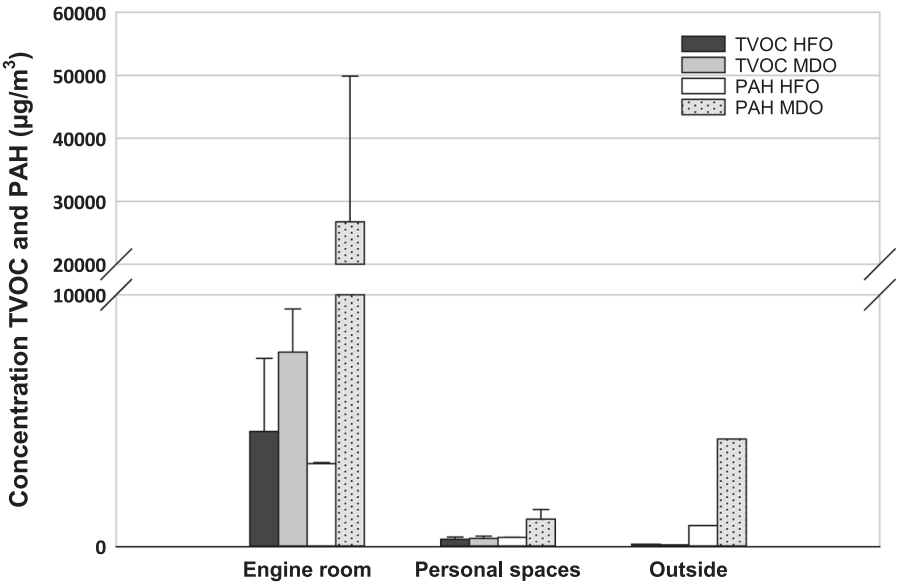

Fig. 3. Concentration of TVOC and PAHs measured in the ship compartments and outside. The error bars are 95%-confidence intervals except for the outside measurement and the PAHs during HFO-campaign (single points).

campaign reflects the higher proportion of these PAHs in the MDO than HFO fuel. TVOC and the 32 PAHs were substantially higher in the engine room compared to the personal spaces (Fig. 3).

3.4. Particles – Concentrations and size distributions

$PM_{2.5}$ concentrations and particle number concentrations of nanoparticles (PNC; particle diameter 10–1000 nm) from the individual measurement sites for both fuels are presented in Table S9. The medians of particles, both $PM_{2.5}$ masses and the PCN, were statistically significantly lower ($p < 0.05$) in the MDO case (Table S2). Detailed comparison of concentration in the engine compartments does not show systematic differences; $PM_{2.5}$ being increased by a factor of two in the purifier room but similar in the boiler room while PNC increased by a factor of five in the boiler room and was similar in the purifier room. The systematically decreased concentrations in the MDO case in the personal compartments could be caused by the lower influence of exhaust entering by the ventilation. However, on the averages for the compartments, there was a decrease in $PM_{2.5}$ and PNC for the MDO fuel even if the personal spaces might have been affected by cleaner outside air incoming with the ventilation flows (Fig. 4).

The size distribution of nanoparticles differed slightly between the fuels, for the particles in the boiler and purifier rooms and the

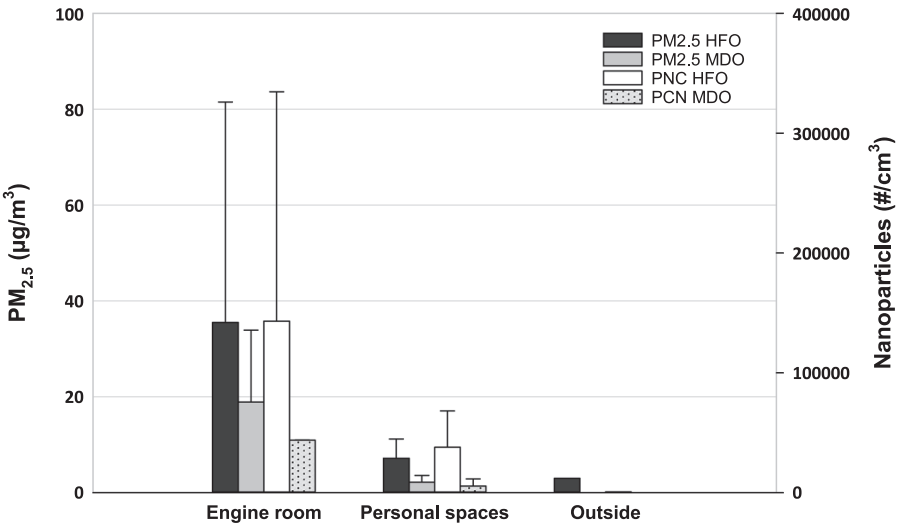

Fig. 4. Concentration of $PM_{2.5}$ and number concentration of nanoparticles measured in the ship compartments and outside. The error bars are 95%-confidence intervals except for the outside measurement (single point).

engine office, moving the mode to higher sizes: 10–30 nm for HFO and 50–70 nm for MDO (Figs. S5–S7). The nanoparticle size distributions at the other sites then the engine space remained unchanged (in Figs. S8–S13).

3.5. Chemical composition of $PM_{2.5}$

The chemical composition of $PM_{2.5}$ was determined from the results of the analyses of EC and OC in particles collected on quartz filters and elements in particles collected on PTFE filters. The concentrations of EC, OC and the elements are shown in Table S10. The elements selected for further analysis were those associated with the ship fuels and lubricants: S, V, Ni, Fe, Ca, Na and Zn; Na and Cl with sea salt and Al and Si with mineral dust. To calculate the relative contribution of the different compounds to $PM_{2.5}$ mass, sulfur in the particles is expressed as sulfate SO_4^{2-} and sulfate-bound water is calculated as sulfate × 0.8, similarly as in Moldanová et al. (2013). Organic carbon is expressed as organic mass (OM), calculated as OM = 1.2 × OC (Petzold et al., 2008). Sum of V, Ni, Fe, Ca, Zn, Na, Cl, Al and Si is denoted as Other elements. The resulted composition of $PM_{2.5}$ collected in the on-board spaces and outside during the two cruises is shown in Figs. S14 and S15. The $PM_{2.5}$ particles consisted mainly of OC and sulfate, even if the fraction of the "other elements" was quite large in some places such as engine control room and engine office. The mass closure of our measurements may be associated with large uncertainties due to sampling on different kinds of filters (quartz for EC/OC and PTFE for the elements and $PM_{2.5}$ mass). The undefined fraction was slightly larger for the samples collected during the cruise with MDO. The only statistically significant difference ($p < 0.05$) was the fraction of sulfate in the $PM_{2.5}$, consistent with the content of sulfur in the fuels.

Statistically significant differences were observed for decreased concentration of S, Ni, V and Na ($p < 0.05$). There was no statistically significant difference for the concentrations of EC ($p = 0.33$) between the two fuels qualities but the concentration of OC decreased significantly ($p < 0.05$). The exhaust measurements typically show large decrease in emissions of S, Ni and V, all associated with high-sulphur HFO, for the MDO fuel, while differences in EC and OC are not as pronounced (Moldanová et al., 2013).

3.6. Principal component analysis

To describe and illustrate the dominant correlations between the 25 variables measured and the 21 observation sites, all median concentrations of the reported indoor air pollutants and indoor climate parameters were entered into a two component principal component analysis (PCA) model for overview, using the SIMCA software (Umetrics SIMCA). All variables were centered and scaled to unit variance prior to the component computations. There were no outliers in the components, nor in the residuals. (Prior log-transformation of the variables before the computations did not improve the interpretability of the data model). The model explained 53% of the total variation, considered enough for an overview. As seen in the score plot (Fig. 5), there is an apparent difference in the second component for the IEQ when the ship is operating on heavy fuel oil (HFO in red) and on marine diesel oil (MDO in green). The first principal component is dominated by the variation of the ventilation rate (air exchange rate, AER, c.f. Table S3 and assuming no substantial changes in the AER during the MDO campaign) between the measurement sites on the ships and the second component is dominated by the fuel type used.

The loading plot of the model (Fig. 6), illustrates the correlation between the variables. The high covariation of formaldehyde and

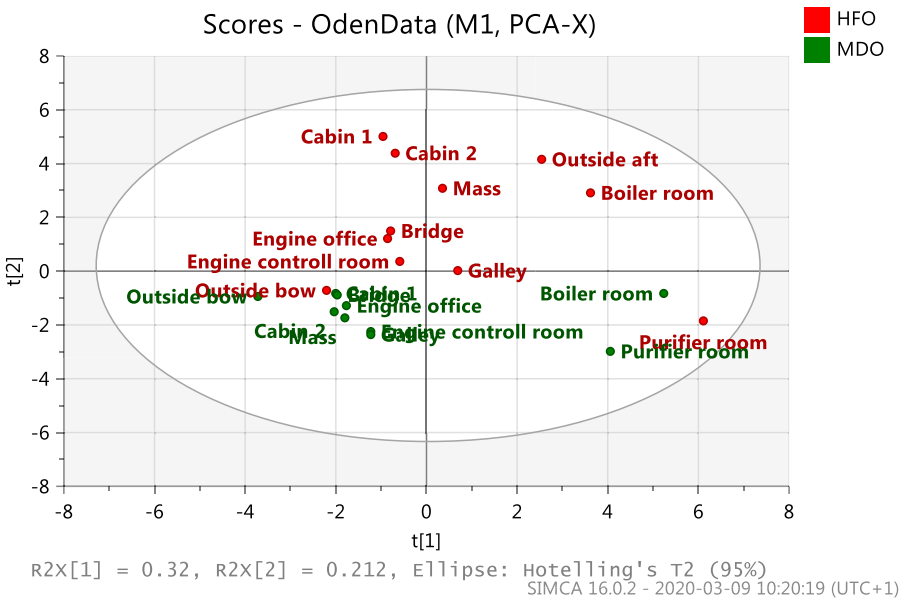

Fig. 5. Score plot from the PCA, showing the correlation between the observation sites, colored by fuel type HFO (red), MDO (green). The spread along the X-axis, first component, can be considered as related to ventilation rate, and the Y-axis, second component, spread is related to fuel type. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

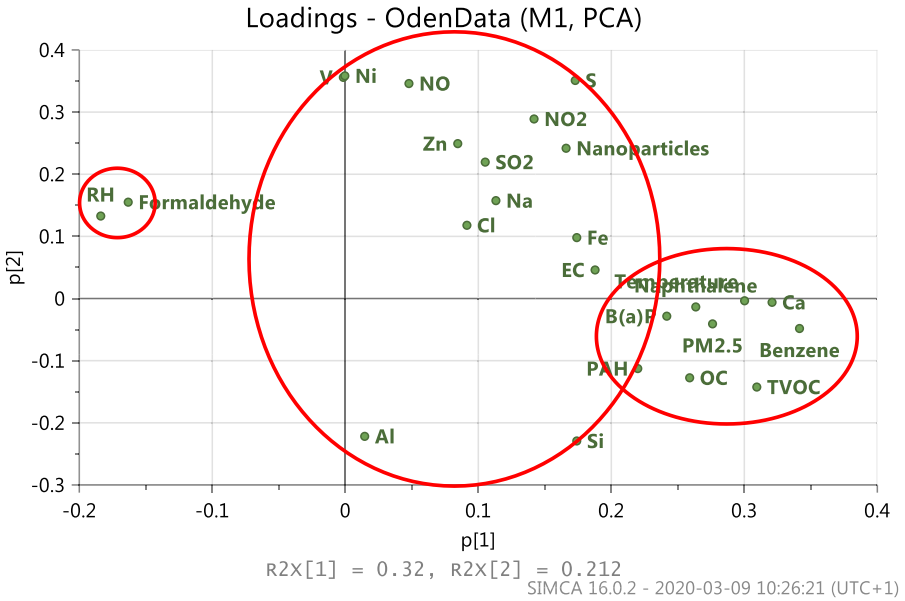

Fig. 6. Loading plot from the same PCA showing the relationship between the variables measured. The variables can be considered clustered into three groups.

RH was also observed for building materials (Huang et al., 2016). Elements from fuel are grouped together with NOx, SO_2, nanoparticles and EC, typical components in combustion exhaust gases. The third group of variables contains mostly parameters associated with evaporative emissions.

4. Conclusions

To summarize, the results show the impact of the fuel on the indoor environmental quality on-board a ship when operating on HFO and MDO, respectively. The engine room areas are characterized by high temperature, low relative humidity and high concentrations of pollutants related to evaporative emissions from fuels, i.e. VOCs and PAHs as well as nanoparticles, NOx and in the HFO case also SO_2. The personal spaces affected by ventilation/air exchange with outside air show lower temperatures, higher relative humidity and low concentrations of VOCs and PAHs. Concentrations of NO and NO_2 reveal that ventilation system might be affected by exhaust emissions. These emissions can enter the accommodation via ventilation system from for example the exhaust funnel or a tank goose-neck ventilator. The prevailing northern wind direction, relative to the ship's movement, in both campaigns, indicates similar extent of the indoor air pollution due to intake of outside air polluted by the ship's engine exhaust.

We acknowledge that the campaigns were carried out in different conditions regarding the lapse of time (3 years between the campaigns), mode of operation of the ship and external environmental conditions. These conditions were harsher for the MDO campaign: operating in ice with associated higher engine loads, lower outdoor temperature and higher relative humidity. Even though, we could observe an improvement of the indoor air quality on the ship while operating on the MDO fuel.

The largest differences were observed for the median concentrations of SO_2 and NO_2, and PM constituents sulphate, V and Ni associated with the HFO fuel. PAH levels were, however, higher during MDO operations, indicating that elevated levels of alkylated PAHs in the fuel affect the IEQ on-board. The concentrations of all air pollutants were mostly below relevant indoor air guideline values and far below occupational exposure limit values. Hence, this kind of intervention, primarily intended to protect the outdoor environment is also beneficial for the indoor environment on-board a ship. In extension, it has implications for the personal exposure and thus the health and well-being of the crew, both in terms of reduced exposure to harmful air pollutants, but also in reduced dermal contact to HFO during service and cleaning tasks. The study shows that a policy measure (the SECA regulation) to protect ambient environment from ship emissions brings unintentional, yet important benefits for the indoor environmental quality onboard a ship. The study emphasizes the need to further investigate the impact of new marine fuels, their chemical composition and technologies from a system perspective, including effects on workers' occupational health and safety.

CRediT authorship contribution statement

Sarka Langer: Conceptualization, Funding acquisition, Investigation, Writing - original draft. **Cecilia Österman:** Funding acquisition, Investigation, Writing - original draft. **Bo Strandberg:** Formal analysis, Writing - original draft. **Jana Moldanová:** Methodology, Writing - original draft. **Håkan Fridén:** Formal analysis.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.trd.2020.102352.

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